JOURNAL OF GEOPHYSICAL RESEARCH, VOL. 101, NO. D2, PAGES 3901–3920, FEBRUARY 20, 1996

Major optical depth perturbations to the stratosphere from volcanic eruptions: Pyrheliometric period, 1881–1960

Richard B. Stothers

Institute for Space Studies, Goddard Space Flight Center, NASA, New York

Abstract. A detailed chronology of major stratospheric dust veils from 1881 to 1960 has been constructed by searching the primary literature for relevant observational data of various kinds, especially pyrheliometry. Data from 23 observing sites in both hemispheres have been reduced in a rigorous fashion to yield a table of pyrheliometric optical depth perturbations as a function of year, month, and latitude band. To convert to visual reference wavelengths, the tabular entries should be multiplied by a time-dependent factor that is somewhat greater than unity. Ten measurable volcanic dust veils have been established in this manner and have been analyzed as to their generation, transport, decay, and total mass. These clouds arose from eruptions of Krakatau (1883), an unidentified volcano (1890), Soufrière and Pelée (1902), Santa Maria (1902), Ksudach (1907), Katmai (1912), Puyehue (1921), Paluweh (1928), Komagatake (1929), and Quizapu (1932). Other turbidity indicators have also been used in the analysis, including starlight extinction, purple twilight glows, color of Sun and Moon, polarization of blue sky light, Bishop's ring around the Sun, and dark lunar eclipses. Global stratospheric aerosol loadings have been computed from the peak turbidities. Agreement with the aerosol masses derived from polar ice cores is good in the important cases of Krakatau, Santa Maria, and (after a major correction of the ice core value) Katmai. During the long period 1881–1992, about 80% of all stratospheric aerosols generated by the largest sulfur-producing eruptions were emplaced during the two short time intervals 1883–1902 and 1982–1991. The long-term (1881–1992) average annual production rate of stratospheric SO_2 from the largest eruptions was 0.8 Tg yr^{-1}, about half the average rate since 1981. Implications of the principal results are discussed.

1. Introduction

When volcanoes explode, the fiery colors in the sky sometimes linger long after the eruption has ceased. In a stratosphere-penetrating event the erupted sulfur gases (mostly SO_2 and H_2S) rapidly combine with water vapor in a complex series of chemical reactions to form long-lasting aerosols of approximately 75% H_2SO_4 and 25% H_2O [*Toon and Pollack*, 1973; *Castleman et al.*, 1973]. These and the directly injected sulfur aerosols, along with some small silicate ash particles, constitute what is known as a "dust veil." The cloud of particles is transported rapidly around the globe by stratospheric zonal winds, while slowly spreading both zonally and meridionally. Although the larger ash particles gravitationally settle out of the stratosphere within a few days or weeks, and are then quickly removed to the ground by rainout, the small sulfate droplets (and some tiny ash specks) remain suspended for months to a few years. They give rise to vividly colored sunsets and other kinds of optical phenomena that are visible for a long time over a wide band of latitude [*Symons*, 1888]. When the aerosol particles themselves eventually fall out, they can be indirectly detected years afterward as acid contaminants in glacial ice [*Hammer*, 1977].

A primary aim of studying these diverse phenomena is to determine the spatial and temporal distribution of volcanically produced stratospheric turbidity, which is a necessary ingredient in precise modeling of the climate system. The most useful measurements for the period between 1881 and 1960 are those that were made by pyrheliometers. Despite many efforts since 1837, all early pyrheliometers remained inadequately calibrated and unsystematically deployed until the celebrated Montpellier series of direct-beam observations of the Sun in 1882–1900 [*Eon*, 1901] and the less accurate Berlin series in 1881–1886 [*Frölich*, 1887]. Ironically, none of the measurements made before 1902 was originally intended to look for volcanic perturbations to the atmosphere; rather, their only purpose was to determine the "solar constant" and its possible variability. *Pernter* [1889] and *Abbot* [1902], however, noted the usefulness of such measurements for volcanic studies.

Beginning in 1910, many authors constructed long-term composite time curves of solar radiation intensity by using the monthly observations supplied by a number of United States and other northern midlatitude stations [*Kimball*, 1918, 1924; *Hand*, 1939; *Hoyt et al.*, 1980]. Kimball's data have been heavily exploited by *Pollack et al.* [1976] and by *Sato et al.* [1993] in their derivation of mean monthly optical depth perturbations due to volcanic eruptions since 1883. However, Kimball used only 13 stations and did not make any corrections of the published intensities for the monthly changes of local noontime air mass. He also adopted a reference baseline that included both volcanically disturbed and volcanically undisturbed years and applied a smoothing filter (three-point averaging) before plotting his monthly data points. Users of Kimball's plots have had to make a rough statistical correction of the baseline and have had to prune the data by excising 10 months of the year for

Paper number 95JD03237.

certain stations in order to minimize the air mass problem. Although *Dyer* [1974] reanalyzed some of the original radiation data in a more rigorous fashion, he chose only four stations. In still another approach, *Bryson and Goodman* [1980] derived absolute solar intensities from the original data by attempting to correct for all aerosol and nonaerosol constituents of the atmosphere, thereby avoiding the need to use reference years. This method, however, is not reliable except for stations with exceptionally clear and stable atmospheres, as *Abbot et al.* [1932] have shown. Very few of the 42 stations used by Bryson and Goodman satisfy these demanding requirements. Even then, adoption of the reference year method is still desirable [*Hoyt*, 1979a]. In any case, Bryson and Goodman's final results have been displayed only as annual averages of aerosol optical depth over the northern midlatitudes. All of the foregoing authors assumed that pyrheliometric and visual optical depths are approximately equal, whereas in reality the visual optical depth can often exceed the pyrheliometric by a significant amount (section 2.2).

Using, in part, atmospheric transmission data but also, more extensively, measurements of the total ejected tephra volume and, to a lesser extent, tropospheric temperature decreases after large volcanic eruptions, *Lamb* [1970] constructed a "dust veil index" (dvi) for many eruptions in the period 1500–1968. Though acknowledging the obvious crudeness and indirectness of a tephra-based approach, *Mitchell* [1970], *Oliver* [1976], and *Robock* [1981] employed it, wholly or partly, as a provisional tool to create their own time series of annual volcanic dust loadings of the stratosphere since the years 1850, 1883, and 1600, respectively.

Relatively speaking, the most reliable of all these time series is probably the one published by *Sato et al.* [1993]. It may be in fact almost the best that can be put together by using only secondary sources of pyrheliometric data for the period 1881–1960. A clear advance, however, could obviously be made by returning to the primary literature, searching for all types of pertinent observational data, and making critical selections and careful reductions of the most valuable data in the light of modern knowledge. The present author has already applied this exhaustive, historical type of approach to investigations of the great dust veils thrown up by Tambora (1815), Laki (1783), and the mystery volcano of AD 536 [*Stothers*, 1984a, b, 1995]. Because the primary literature searches and data reductions were all carried out in the same way by the same person, a large degree of homogeneity in the results was achieved.

In the present paper, the same approach has been used for all known major volcanic eruptions between 1881 and 1960. Since the most important perturbations to the stratosphere during this long period occurred during the first half of it, the search of the primary literature was focused most intensively on the years before 1920. A brief conspectus of the paper follows. Section 2 discusses the pyrheliometric aspects, specifically, how the measured solar radiation intensities were reduced, what data were finally adopted, and what constant was used to convert from pyrheliometric turbidity to visual turbidity. Section 3 describes six other methods that were used to quantify (or at least to recognize) stratospheric turbidity: extinction of starlight, twilight glows, color of Sun and Moon, polarization of blue sky light, Bishop's ring around the Sun, and dark lunar eclipses. Section 4 provides a chronological account of the generation, transport, and decay of each stratospheric dust veil, volcano by volcano. Section 5 presents a compilation and analysis of the final body of turbidity results, in the form of a chronological table of monthly mean optical depth perturbations as a function of latitude band. In section 6 the masses of the dust veils are calculated from the inferred turbidities, while in section 7 the mean annual stratospheric sulfur output from the largest eruptions is computed and compared with the estimated outputs for other time periods. Section 8 summarizes the main results and concludes the paper.

2. Pyrheliometry

2.1. Reduction of the Pyrheliometric Data

A solar beam of intensity I_0 impinging on the top of the Earth's atmosphere attenuates downward in accordance with *Bouguer*'s [1729] law,

$$I = I_0(e^{-\tau})^m \tag{1}$$

where m is the air mass in the direction of the Sun. In astronomical work the optical depth, τ, is usually replaced by the (decimal based) astronomical extinction in magnitudes,

$$a = [2.5 \log_{10} e]\tau \approx 1.086\tau. \tag{2}$$

All sources of absorption and scattering are implicitly included in τ. If a distinction is made between the unperturbed (or normal) contribution, τ_N, and the volcanic contribution, τ_D, then $\tau = \tau_N + \tau_D$. It is still the standard practice to compare observations of the cloud-free Sun at the same air mass and at the same time of the year, during the unperturbed and the volcanically perturbed periods, in order to evaluate τ_D at any given site [*Kimball*, 1918]. In doing so, one circumvents the problems of (1) air mass differences at different times of day and at different times of year, (2) the changing Earth-Sun distance, (3) seasonal variations of the normal extinction, τ_N, and (4) various other corrections, such as that for altitude of the observing site. It must be assumed that τ_N remains unchanged between the normal and the perturbed periods, in particular, that the atmospheric water vapor content on clear days is not significantly changed, but many studies have shown that this is a quite good assumption. Under the stipulated protocol, (1) leads to

$$\tau_D = m^{-1} \ln (I_N/I_D) \tag{3}$$

where I_N refers to the normal period and I_D to the perturbed period. If I_D remains fairly close to I_N, (3) becomes, approximately,

$$\tau_D = m^{-1}(I_N - I_D)/I_N. \tag{4}$$

A further simplification is possible if the solar zenith angle, z, is sufficiently less than 90° that the height of the volcanic perturbing layer need not be entered into the calculation and atmospheric refraction, too, can be ignored. The air mass in that case is

$$m = \sec z. \tag{5}$$

Since all of the reductions performed in the present paper apply to $z < 83°$, any errors in m due to our use of this approximation should be less than 5%.

To reduce the effect of day-to-day turbidity fluctuations, monthly means of radiation intensities (or of the atmospheric transmissions) are formed. Days used here include only those on which measurements were obtained for the cloud-free Sun within one hour before or one hour after local noon. This procedure essentially eliminates the influence of systematic

Table 1. Stations With Pyrheliometric Data Subjected to New Reduction

Station	Country	Lat	Long	Period	Sources of Data
Clarens & Lausanne	Switzerland	46°N	7°E	1896–1904	*Dufour and Bührer* [1903], *Bührer and Dufour* [1905]
Davos	Switzerland	47°N	10°E	1912–1926	*Lindholm* [1927]
Helwan	Egypt	30°N	31°E	1914–1921	*Knox-Shaw* [1923a, b]
Kew	England	51°N	0°W	1908–1921	*Kew Observatory* [1909–1911], *Watson* [1923]
Lincoln, Nebraska	United States	41°N	97°W	1910–1915	*Kimball* [1916]
Madison, Wisconsin	United States	43°N	89°W	1910–1917	*Kimball and Miller* [1912, 1916], *Kimball* [1916, 1917], *Hand* [1917]
Madrid	Spain	40°N	4°W	1910–1920	*Madrid Observatory* [1911–1920]
Monte Cimone	Italy	44°N	11°E	1902–1905	*Chistoni* [1906]
Monte Rosa	Italy	46°N	8°E	1905–1907	*Alessandri* [1908]
Montpellier	France	44°N	4°E	1882–1900	*Eon* [1901]
Mount Brukkaros	Namibia	26°S	18°E	1926–1930	*Abbot et al.* [1932]
Mount Montezuma	Chile	23°S	69°W	1920–1925	*Abbot et al.* [1932]
Mount Stromlo	Australia	35°S	149°E	1931–1936	*Rimmer* [1937]
Mount Weather, Virginia	United States	39°N	78°W	1907–1914	*Kimball* [1913a, 1914a, b]
Mount Wilson, California	United States	34°N	118°W	1905–1920	*Abbot and Fowle* [1908], *Abbot et al.* [1913, 1922]
Paris	France	49°N	2°E	1907–1914	*Angot* [1911–1913], *Dufour* [1915–1920]
Pavlovsk	Russia	60°N	30°E	1892–1913	*Savinov* [1913]
				1912–1922	*Kalitin* [1926]
Potsdam	Germany	52°N	13°E	1902–1915	*Scheiner* [1908]; *Martin* [1913, 1914], *Süring* [1915, 1917]
Santa Fe, New Mexico	United States	36°N	106°W	1910–1917	*Kimball* [1915, 1916, 1917], *Hand* [1917]
Simla	India	31°N	77°E	1906–1916	*Walker* [1908–1918]
Uppsala	Sweden	60°N	18°E	1909–1922	*Sjöström* [1930]
Warsaw	Poland	52°N	21°E	1901–1918	*Gorczyński* [1913], *Stenz* [1919]
Washington, D. C.	United States	39°N	77°W	1902–1907	*Abbot and Fowle* [1908]
				1905–1912	*Kimball* [1910, 1913a]

differences between morning and afternoon measurements that exist at some stations. A second advantage is conferred by the minimal path length of the beam at noontime, which lessens the influence of unknown atmospheric conditions far from the observing station. We make use of all days in which skies were listed as being clear around the Sun, not just that one day in each month with the highest measured sky transparency. This flexibility seems to make sense: it enlarges the statistical base in the presence of occasional accidents like outright mismeasurements or abnormally high apparent transparencies (for example, those measured from time to time in the clearings between cumulus clouds). Also, many of the tables of radiation data in the published literature do not contain the daily raw data but only the derived monthly means, which we are obliged to accept. In the latter case, the authors' criteria for selecting data are usually not spelled out, and it is safest to assume that the authors used as much of their data as possible. In instances that we could check, they routinely had done so. It is also often the case that authors have presented their data only for one or more selected air masses; if a choice is possible, we adopt the smallest air mass for which sufficient data are available to form useful monthly averages.

In the category of volcanically disturbed years we place the following eighteen: 1883–1886 (Krakatau), 1890–1892 (unknown volcano), 1902–1904 (Soufrière, Pelée, Santa Maria), 1907–1908 (Ksudach), 1912–1914 (Katmai), 1928–1929 (Paluweh, Komagatake), and 1932 (Quizapu). The years 1921–1922 (Puyehue) were only slightly disturbed and so are not listed. All other years between 1881 and 1960 (whenever published data exist for them) are regarded as essentially undisturbed in forming the normal monthly means for each observing station. If a station record is very short, data up to the month of the eruption in an eruption year are also averaged into the station norms. Any months in which local dust storms or local forest fires are known to have occurred are omitted for the affected station.

Calculation of the optical depth perturbations follows from (3). In the case of the long-operating Swiss station at Davos, *Lindholm* [1927] has already published monthly mean values of τ_D, by using an approximation equivalent to (4).

Computed values of τ_D will occasionally be slightly negative during periods of low volcanic activity. This is an inevitable consequence of the reference year method. Though such values are unphysical, they do provide a useful measure of the accidental background scatter around the monthly norms for each station. Therefore they allow an assessment of the reality of any apparent positive departures that could indicate a volcanic perturbation. When uncorrelated results for several stations are averaged, the results should yield a close approximation to the true monthly value of τ_D.

Pyrheliometric data that have been used in this paper were obtained only by standard pyrheliometers. Data from other types of measuring devices, such as pyranometers, black and bright bulb actinometers, and sunshine duration recorders, have proven to be too difficult to calibrate accurately, although they are sometimes useful for at least detecting the existence of an optical depth perturbation. Stations for which the accepted pyrheliometric data have been formally reduced in this paper are listed in Table 1, where the sources of data are also cited. The specified time windows usually represent the full record lengths if they are short; in other cases the records have been truncated in order to minimize the effects of possible small changes occurring in the local background turbidity. Potential background problems are the increases of manmade pollution and the spurious effects engendered by degradation, upgrading, relocation, and replacement of the station pyrheliometers. Note the concentration of stations at latitudes between 30°N and 60°N.

For stations [*Kimball*, 1927, 1930] not listed here, one or more explanations for their omission may apply. In some cases the data series are too sparse, too short, or too erratic to obtain accurate normal monthly means. In other cases the series do not cover enough of a volcanically perturbed period to be relevant. In general, data from Russia and the former Soviet Union (except for Pavlovsk) have not been used, as coverage in the northern midlatitudes is already adequate; *Pivovarova* [1970], however, has displayed pyrheliometric data from eight former Soviet stations. In cases where the volcanic perturbations are small enough to be neglected, as during the 1940s and 1950s, we have simply accepted the results of earlier published reductions and analyses of the observational data. For the eruptions of Paluweh (1928), Komagatake (1929), and Quizapu (1932) we have made full use of *Hoyt*'s [1979a] analyses of the extensive data secured at the Mount Montezuma (Chile) and Table Mountain (California) stations by staff members of the Astrophysical Observatory of the Smithsonian Institution (APO).

For northern midlatitudes, temporal coverage is complete over the whole period of this study, 1881–1960. However, no single pyrheliometric station covers the entire period.

2.2. Conversion Between Pyrheliometric and Visual Turbidities

Pyrheliometers respond about equally to radiation of all wavelengths at least between 0.3 μm and 2 μm, i.e., over the bulk of the solar spectrum which is accessible from the ground. However, a reference interval used frequently in theoretical climate studies is the visual band centered around 0.55 μm. In only a very few cases are spectrobolometric measurements available from which the visual response can be directly obtained. Therefore it is advantageous to use the large mass of available pyrheliometric observations to determine a pyrheliometric turbidity, which can then be converted to a visual turbidity by multiplying by an appropriate factor, k.

The value of k has been variously estimated. *Dyer and Hicks* [1968] simply took $k = 1$, which would apply if the volcanically produced particle extinction were independent of wavelength, λ. This might be roughly true immediately after the volcanic eruption, when large particles of ash dominate the dust cloud before they rapidly drop out of the atmosphere. In the ensuing weeks, however, the extinction is observed to progressively deviate from its approximate neutrality, at shorter and shorter wavelengths.

Volz [1970, 1975a] showed that only a month or two after the large eruptions of Katmai (1912) and of Agung (1963), the extinction was already following a λ^{-1} dependence for wavelengths from 0.4 μm out into the infrared. Now the effective wavelength of solar radiation is

$$\lambda_{\text{eff}} = \int_0^\infty \lambda F_\lambda \, d\lambda \Big/ \int_0^\infty F_\lambda \, d\lambda \tag{6}$$

where F_λ is the solar irradiance at the top of the Earth's atmosphere. Use of *Allen*'s [1973] tabulated data for the Sun yields $\lambda_{\text{eff}} = 0.9$ μm. Therefore as Volz originally argued, $k = (0.55\ \mu\text{m}/0.9\ \mu\text{m})^{-1} = 1.6$.

Similarly, *Langley* [1903] and *Abbot* [1903] have published spectrobolometrically obtained transmission data for the year after Santa Maria's (1902) eruption, from Washington, D. C., observations. The extinction followed a λ^{-1} dependence, and the ratio of the measured extinction at 0.5–0.6 μm to that at 0.9–1.0 μm was 1.6.

We emphasize that these three independent estimates of k from observations made after the eruptions of Katmai, Agung, and Santa Maria utilize the fact that pyrheliometers absorb at nearly the same effective wavelength as the effective wavelength at which the Sun emits.

An independent and better way to estimate k is to directly compare simultaneously obtained pyrheliometric and spectrobolometric measurements of the aerosol extinction. Data of this kind were used by *Volz* [1975a] to calculate k for the Katmai dust veil one month after the eruption. From observations made at Mount Wilson, California, and at Bassour, Algeria [*Abbot et al.*, 1922], Volz obtained $k = 1.7$.

Volz [1970] found it necessary to use a roundabout method for the period following the Agung eruption. Instead of solar spectrobolometric measurements that were not available, he used visual measurements of the extinction of starlight (from a variety of stations at 30°–40°S) and compared them with a set of pyrheliometric extinction measurements made at Aspendale (38°S), Australia [*Dyer and Hicks*, 1965]. He thus found $k = 1.7$.

A few simultaneous measurements have been secured with a pyrheliometer and a spectrobolometer at Washington, D. C., between October 1902 and May 1907 [*Abbot and Fowle*, 1908]. However, the only months with sufficiently numerous data of good to excellent quality are February and March of those years. Two acceptable days in 1903, compared in regard to their extinction data with five acceptable days in the normal period 1904–1907, yield $k = 1.4$, about four months after the Santa Maria eruption.

To conclude this section, we have found much support for Volz's contention that the correct value of k to use is approximately 1.6, that this value is to be taken as constant in time (for a while) after the first few weeks following the eruption and that it probably applies to all eruptions. Because pyrheliometers and spectrobolometers admit into their entrance apertures some scattered sunlight in addition to the direct solar beam, a correction for forward scattering by the aerosols is necessary. For wavelengths longer than about 0.4 μm the correction is found to be less than 1% for typical instruments with angular apertures smaller than 10° [*Abbot et al.*, 1932; *Sutherland et al.*, 1975]. Such a small correction can be neglected. Moreover, the ratio, k, of the turbidities will be affected much less than are the visual and pyrheliometric turbidities themselves. Therefore in this paper we adopt the relation

$$\tau_{\text{vis}} = 1.6\tau_{\text{pyr}}. \tag{7}$$

This relation, however, can only hold if most of the aerosols are as small as indicated by the λ^{-1} dependence of the turbidity over the directly measured range 0.4 μm $\leq \lambda \leq$ 2 μm [*Volz*, 1970; *Deirmendjian*, 1973; *Toon and Pollack*, 1976]. Mie scattering calculations suggest that the effective radius, r_e, of such aerosols would be about 0.3 μm [*Lacis and Mishchenko*, 1995]. Since the detailed time histories of r_e and hence of k after the

Table 2. Greatest Diminutions of the Monthly Mean Percentage of Blue Sky Polarization Measured at the Point of Maximum Polarization

Year	Month	Site	Normal, %	Disturbed, %	Source
1884		Paris	58[a]	37[a]	*Cornu* [1885]
1903	June	Washington, D. C.	60	38	*Kimball* [1910]
1907	July	Washington, D. C.	62	53	*Kimball* [1910]
1912	August	Mount Weather, Virginia	59	32	*Kimball* [1913a]

[a]Monthly maximum percentage times 0.78, where the approximate conversion factor is derived from *Kimball* [1913a].

first few months following the eruptions discussed here are not known and the history of r_e for even the well-observed Pinatubo (1991) eruption depends largely and unaccountably on the method of measurement [*Russell et al.*, 1993; *Asano*, 1993; *Sassen et al.*, 1994; *Rosen et al.*, 1994], we simply assume (7) for all subsequent times as a temporary expedient. At least in the months right after the eruptions of El Chichón (1982) [*DeLuisi et al.*, 1983] and Pinatubo [*Russell et al.*, 1993] the wavelength dependence of the turbidity was roughly the same as that measured after the other eruptions.

3. Other Turbidity Indicators

3.1. Extinction of Starlight

Starlight can be used as a probe of the upper atmosphere in the same manner as is the light of the Sun. In the case of the fixed stars, however, the published intensities are conventionally reduced to zero air mass and the total extinction is calculated by standard methods based on (1), (2), and (5).

Before the late 1950s such extinction data were not routinely saved [*Laulainen*, 1977], and in the period of greatest interest here, which runs from 1883 to 1914, they were not even acquired photoelectrically and were only rarely determined photographically. Stellar magnitudes of that period and earlier are so poorly determined that measurement errors of several tenths of a magnitude are common even in relative photographic photometry. Unless a star of known magnitude happens to have been observed at a large zenith angle, where its desired total zenithal extinction would be multiplied by a large factor (sec z), any attempt now would be fruitless to extract from the estimated brightness of the star a possible excess zenithal extinction of several tenths of a magnitude, which is characteristic of major early 20th century volcanic dust veils. For these reasons, stellar extinction data, though diligently searched for, have only marginal utility in this paper.

3.2. Twilight Glows and Colored Sun and Moon

If the optical thickness of the spreading dust cloud exceeds a minimum value, yet is not too large, brilliant purple and red glows decorate the posteruption twilight sky. Their duration provides a useful measure of the height of the glow stratum [*Symons*, 1888]. Since it is otherwise hard to describe and quantify these subtle phenomena, they have not yet provided an accurate index of the dust cloud's optical thickness [*Volz*, 1970, 1975a; *Deirmendjian*, 1973; *Coulson*, 1988].

Data from the present paper, however, can be used to determine at least the critical optical thickness above which the glows disappear. After the eruption of Santa Maria the glows were seen only feebly and infrequently between February and August 1903. Similarly, after Katmai, they seemed to disappear between mid-June and late September 1912. At all four of these turning points the pyrheliometric optical depth perturbation was very nearly 0.09 (visually, 0.15).

The glows also were suppressed after the great eruptions of Laki (1783) and Krakatau (1883), but not enough detailed information is available about the specific times and optical depth perturbations in these two cases to accurately calculate the critical optical thickness. In the case of Tambora (1815), the brilliant glows were extinguished for a very long time, between late September 1815 and July 1818 [*Howard*, 1833]. At the latter date, the visual optical depth perturbation is estimated to have been 0.17 [*Stothers*, 1984a]. Such close agreement with the expected value of 0.15 provides additional confirmation that the scale of the visual extinction curve published for the Tambora dust cloud is essentially correct.

If the optical thickness of the dust cloud increases beyond a few tenths (the exact threshold is still undetermined), the Sun, Moon, planets, and stars can become unusually colored. Most often, the conferred color is green or blue [*Deirmendjian*, 1973]. A still greater optical thickness renders all the luminaries reddish or, eventually, whitish [*Stothers*, 1995]; the necessary optical thickness is probably ~2. However, these numbers depend to some extent on the composition (aerosol or ash) and size distribution of the particles.

3.3. Polarization of Blue Sky Light

Cornu [1885] noticed two striking anomalies as a result of his visual measurements of the sky polarization in 1884 following the Krakatau eruption. First of all, the percentage of blue sky polarization measured at the point of maximum polarization was much less than values obtained in the immediately preceding years. Since the source of the sky's polarization is known to be primarily Rayleigh scattering by air molecules and by other particles smaller than the wavelength of visual light, the presence of larger particles created by a volcanic eruption can most likely only diminish the observed percentage of sky polarization, p. A simple but reasonable assumption is that p_D during the disturbed period should be related to p_N in normal times by $p_D = p_N \exp(-c\tau_D)$, where c is a constant. One might therefore expect to observe a linear relation,

$$\ln(p_N/p_D) - c\tau_D = 0. \qquad (8)$$

Table 2 lists the measured monthly mean values of p_D at those times when p_D fell to a minimum after four large volcanic eruptions in 1883, 1902, 1907, and 1912. Omitting the 1884 polarization data for which the corresponding optical depth perturbation is unknown, Figure 1 shows $\ln(p_N/p_D)$ plotted against τ_{pyr}, where τ_{pyr} is the monthly mean pyrheliometric optical depth perturbation recorded at stations with latitudes near the latitude of the polarization-measurement site, averaged over the three consecutive months when the percentage

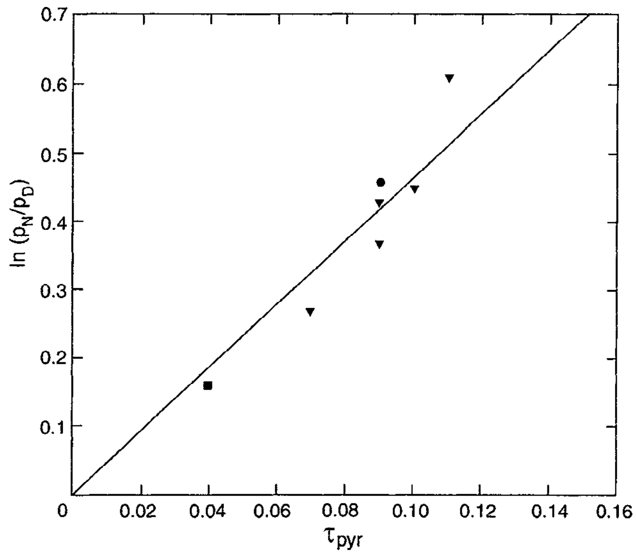

Figure 1. Natural logarithm of the ratio of the percentage of blue sky polarization in normal times to the percentage of blue sky polarization in a volcanically disturbed period, as a function of the pyrheliometric optical depth perturbation. Volcanically disturbed periods shown are June 1903 (circle), July 1907 (square), and August–December 1912 (triangles).

of polarization was least. Also shown are four additional points (based similarly on 3-month averages) corresponding to the four months following the remarkable polarization minimum in 1912. These seven points represent the most reliable data that are available. A regression line having a slope of $c = 4.7$ fits the data in Figure 1 extremely well. This line will be used later to calibrate the 1884 turbidity observations.

The second polarization anomaly that *Cornu* [1885] noticed in 1884 was the shift of the Arago and Babinet neutral points of sky polarization away from their normal positions on the sky. New neutral points also appeared. Although he did not state the amount of the displacements, Busch and many later observers listed by *Jensen* [1928] did so for several eruptions after Krakatau. The average distance of the Arago neutral point from the antisolar point and the average distance of the Babinet neutral point from the Sun, during the normal 12-month period that preceded and during the disturbed 12-month period that followed each eruption in 1890, 1893, 1902, 1907, and 1912, are listed in Table 3 for a true Sun elevation of $-0.5°$, as derived mainly from Busch's results. Although the volcanically caused displacement of the Arago neutral point is relatively small (<3°), the Babinet neutral point moves by 7°, 8°, 16°, 3°, and 6° for the five listed eruptions. Oddly, there appears to be no clear relation between the amount of neutral point displacement and the amount of sky turbidity. However, a very high sky turbidity (as in 1912) is apparently capable of making the neutral point displacement much less than expected.

This paradox does not disappear if other Sun elevations are chosen. Figure 2 shows the run of neutral point position with Sun elevation in the month of peak turbidity after the eruptions of 1902, 1907, and 1912 [*Busch*, 1905, 1908, 1913]; the run during volcanically undisturbed times [*Jensen*, 1937] is also shown. These runs, at least as measured at Arnsberg, Germany, for the three listed eruptions, turn out to have some generality. When the turbidity decreased after the 1912 eruption, the neutral point positions [*Busch*, 1913, 1914; *Jensen*, 1928] shifted through the patterns that are displayed here for the 1902 and 1907 eruptions. Similar curves have been measured at the Mauna Loa Observatory, Hawaii, after the El Chichón (1982) eruption [*Coulson*, 1988]. Although additional factors like aerosol particle size distribution and local low-altitude tropospheric pollution play a small role in establishing the curves [*Neuberger*, 1950], the basic trends are probably secure. The nonmonotonic vertical undulations of the curves with changing turbidity, however, limit the curves' usefulness as a turbidity gauge except possibly at Sun elevations greater than 5°.

Although the slow return of the Arago and Babinet neutral points to their normal positions parallels the gradual decay of the sky turbidity after each eruption, measurable displacements of the neutral points often remain long after the optical depth perturbation can no longer be directly detected [*Kimball*, 1913a; *Volz*, 1969]. Hence sky polarization measurements seem to be the most sensitive indicator of a volcanic perturbation to the stratosphere during the time period covered by this paper.

3.4. Bishop's Ring

After the eruption of Krakatau in 1883, *Bishop* [1884] observed from Hawaii an unusual meteorological halo around the Sun. It consisted of a large diffuse reddish ring surrounding a bright whitish patch. High in the sky the ring appeared essentially circular, but near the horizon it became very distorted. The most careful observations showed that the average

Table 3. Average Displacements of the Arago and Babinet Neutral Points of Blue Sky Polarization at a Solar Altitude of −0.5° During the 12 Months Following Large Volcanic Eruptions

Eruption Year	Site	Arago Neutral Point		Babinet Neutral Point		Source
		Normal	Disturbed	Normal	Disturbed	
1890	Arnsberg	18°	20°	16°	23°	*Busch* [1896]
1893	Arnsberg	18°	20°	16°	24°	*Busch* [1896]
1902	Arnsberg	18°	20°	16°	32°	*Busch* [1903, 1905]
1907	Arnsberg	18°	20°	16°	19°	*Busch* [1908]
1912	Arnsberg	18°	19°	16°	22°	*Busch* [1913, 1914]
	Davos	16°	19°	16°	26°	*Dorno* [1913]
	Mount Weather	19°	19°	17°	21°	*Kimball* [1913a]

Significant displacements also occurred after the Krakatau eruption of 1883 [*Cornu*, 1885].

size of the undistorted ring remained constant in time even after the passage of many months.

Bishop's rings around the Sun (and sometimes around the moon) have been reported after three additional eruptions in the period under present study, in 1890, 1902, and 1912. The average observed radii of the inner and outer borders of the colored ring are listed in Table 4. Although a moderate amount of volcanic dust is needed to produce Bishop's ring, the observed radial dimensions of the ring, which average 12° for the inner edge and 23° for the outer edge, appear to be independent of the total amount of dust, both for the same eruption and for the eruptions of different volcanoes. In recent decades, Bishop's rings of more or less the same size have been reported after the large eruptions of Agung (1963) [*Volz*, 1970], Fuego (1974) and El Chichón (1982) [*Meinel and Meinel*, 1983], and Pinatubo (1991) [*Asano*, 1993; *Sassen et al.*, 1994].

Mie scattering calculations have shown that forward scattering of sunlight by small spherical particles occupying a narrow range of sizes will produce a colored aureole-corona complex of this type. The observed dimensions of Bishop's ring imply that the mean particle radius must be 0.6–0.8 μm [*van de Hulst*, 1957; *Deirmendjian*, 1973; *Asano*, 1993; *Sassen et al.*, 1994]. Even very crude calculations based on first-order diffraction theory for opaque disks give roughly the same result, 0.8–0.9 μm [*Symons*, 1888; *Pernter*, 1889].

Table 4. Average Radii of Bishop's Rings

Year	Inner Radius	Outer Radius	Sources
1883–1884	11°	23°	*Symons* [1888]
1890		~30°	Husmann [*Anonymous*, 1891]
1903	11°	22°	*Forel* [1905]
1912–1913	13°	24°	*Dorno* [1912], *Kimball* [1913b], Schmid [*Maurer and Dorno*, 1914]

These sizes are larger than those implied by the spectral dependence of the turbidity in 1903 and 1912 and may indicate that the size distribution was bimodal. Bimodality or a large size variance could explain the diffuseness of the observed rings.

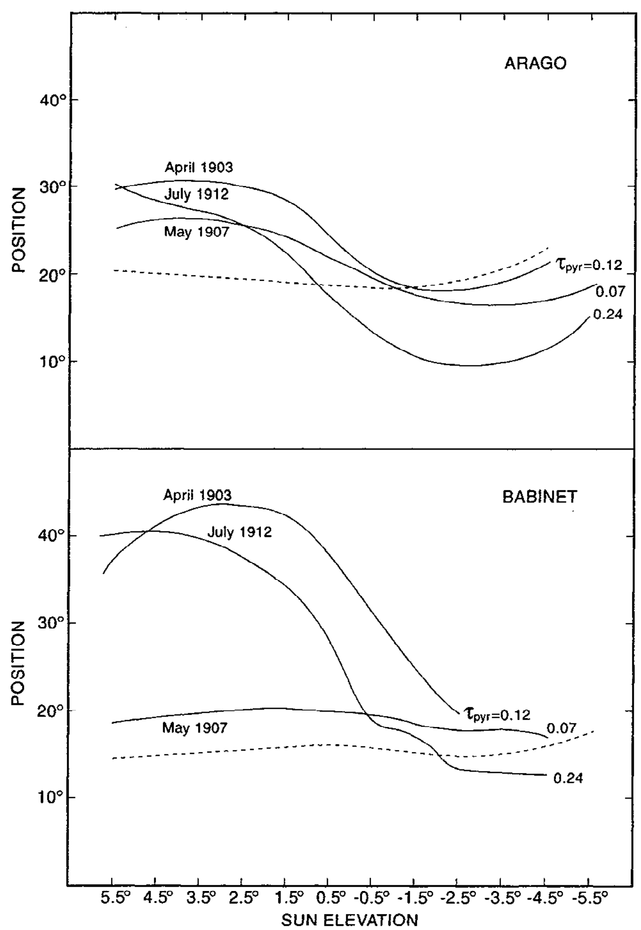

Figure 2. Positions of the Arago and Babinet neutral points of blue sky polarization for various true Sun elevations over Arnsberg, Germany. Solid lines, positions during the period of maximum pyrheliometric optical depth perturbation after three large volcanic eruptions. Dashed lines, undisturbed positions.

3.5. Total Eclipses of the Moon

During a total eclipse of the Moon, rays from the Sun passing tangent to the Earth's atmosphere are refracted and scattered into the Earth's shadow cone and cause the moon to be illuminated with a faint light. If the atmosphere is clear, the moon's surface exhibits a coppery hue, because the red rays are bent more and are also scattered less than the greens and blues. When the high atmosphere contains dust from a large volcanic eruption, the lunar disk appears dark [*Flammarion*, 1884].

Both the amount and the selenographical extent of the lunar darkening provide a crude measure of the optical thickness and geographical coverage of the dust in the Earth's stratosphere. This unique method of probing the dust veil has been most successfully developed in connection with modern observations of dark lunar eclipses [*Link*, 1963; *Hansen and Matsushima*, 1966]. Since, however, the results necessarily suffer from the use of poorer observations, we have made only qualitative use of the method for the early volcanic eruptions discussed in this paper. Nevertheless, a dedicated investigation might be of some interest, especially for earlier times when usable pyrheliometric observations are unavailable. The high frequency of total lunar eclipses (on the average, one per year) makes this technique potentially very useful [*de Vaucouleurs*, 1944; *Keen*, 1983].

4. Volcanic Eruptions

4.1. Krakatau 1883

After several months of minor outbursts the island volcano of Krakatau (6°S), just west of Java, suffered a series of cataclysmic explosions on August 26 and 27, 1883. One stupendous boom that it generated was the loudest sound ever recorded, and the tsunamis unleashed were the highest sea waves volcanically produced in modern times [*Simkin and Fiske*, 1983].

Detailed written accounts of the widespread optical phenomena following Krakatau's eruption have been published by F. A. R. Russell and E. D. Archibald in the "Report of the Krakatoa Committee" [*Symons*, 1888]. White glare in the sky, streaky haze, spectacular sunsets, and an unusually colored Sun and Moon were reported from Krakatau's vicinity immediately after the eruption. These phenomena were followed by prolonged purple and red twilight glows and by Bishop's rings surrounding the Sun. The dust cloud that was responsible for these peculiarities rapidly propagated westward in a narrow zone extending between 33°S and 22°N at an average speed of 35 m s^{-1}. A complete circuit of the Earth required 13 days. Owing to the large optical thickness of the dust cloud the twilight glows and Bishop's rings were usually seen just in front, behind, or at the sides of the main cloud. However, the whitish sky haze and the dim Sun and Moon of a green, blue, silvery, or coppery color were always associated with the dense central portion. The volcanic cloud eventually stretched out longitudinally and spread northward and southward, detaching occasional fragments, until it covered the whole globe by late December 1883. Details of its observed movement have been discussed by *Wexler* [1951b]. The approximately equal rates of fallout of the cloud's aerosols onto the Greenland and Antarctic ice sheets, as determined from modern ice core measurements [*Hammer et al.*, 1980; *Legrand and Delmas*, 1987; *Langway et al.*, 1988], indicate that the stratospheric transport was more or less symmetric between the two hemispheres.

The altitude of the tropical glow stratum in August 1883, derived from the nightly duration of the twilight glows, was about 30 km [*Symons*, 1988; *Pernter*, 1889]. This estimate is consistent with direct evidence of Krakatau's eruption column as having been 30–50 km high [*Verbeek*, 1886]. By January 1884 the main glow layer, now probably due to sulfuric acid aerosols, had sunk to about 17 km, as measured from northern midlatitudes. The colored twilights ended for the most part in April of that year, although they reappeared sporadically with lesser brilliance and shorter duration until late in 1885. Bishop's rings continued to be seen even longer, up to July 1886 [*Symons*, 1888], and sky polarization anomalies persisted until the spring of 1887 [*Busch*, 1889].

Associated with the Krakatau haze was a remarkable 3-year reduction in ground level solar radiation, as *Pernter* [1889] first noted. This anomaly was detected serendipitously by A. Crova at Montpellier in France after the arrival of the main body of dust over Europe in late November 1883 [*Wexler*, 1951a]. The true size of the initial drop, however, cannot be known because of the large monthly fluctuations in the Montpellier observations. Accordingly, the monthly data [*Eon*, 1901] are usually grouped into annual averages and are so displayed here in Figure 3. Annual turbidity rose in 1884, peaked the next year, and fell into the background by 1887. The 1885 average was 0.11. It is difficult to know how accurate this value really is, because, for example, it drops to 0.08 if the two exceptionally high values for August and September are omitted from the annual average. *Crova* [1886] himself attributed most of the high 1885 turbidity to an abnormally large amount of rainfall at Montpellier in that year, with the attendant frequent increase of water vapor in the atmosphere. The Berlin pyrheliometric observations [*Frölich*, 1887] are of no help on this question, because they are too few and too uneven in quality.

Judging from other volcanic eruptions, the peak turbidity over Europe ought to have occurred within a few months after Krakatau's eruption and hence during 1883–1884, not 1885. How can this more reasonable expectation be verified?

The extreme darkness of the totally eclipsed moon on October 4, 1884, is one indication. This lunar eclipse was widely described in the scientific journals of the day, and *Flammarion* [1884] was the first author to connect its darkness to the Krakatau dust cloud. The next eclipse, a partial one, occurred

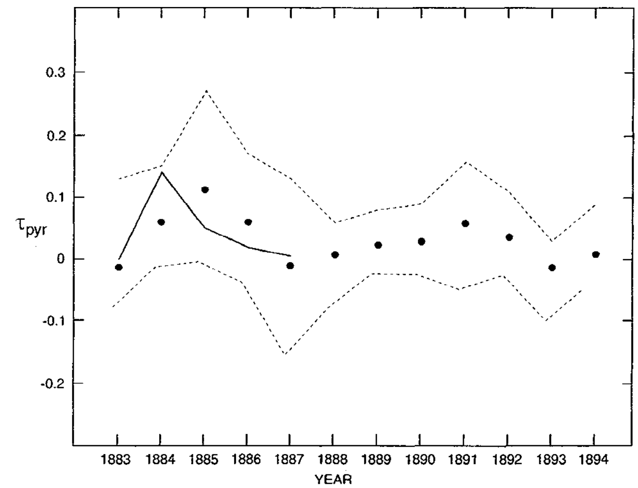

Figure 3. Pyrheliometric optical depth perturbations derived from the Montpellier observations. Circles, annual averages. Dashed lines, largest and smallest monthly mean values in each year. Solid line, adjusted annual averages during the Krakatau period.

on March 30, 1885, and was also very dark within the obscured portion of the moon [*Biggs*, 1885]. However, the partial eclipse of September 23 showed the normal coppery color [*Keeler*, 1885]. This suggests that the average turbidity in 1885 was in fact lower than in 1884.

Another indication that 1884 was the year of peak turbidity comes from modern measurements of volcanically produced sulfate acidity in the annual ice layers laid down at Crête, Greenland [*Hammer et al.*, 1980]. According to a number of independent studies of volcanic (and nonvolcanic) pollution of the stratosphere the fallout of small volcanic particles to the ground at polar latitudes begins to become significant about one year after the original time of injection [*Volz*, 1975b; *Hammer et al.*, 1980; *Fiacco et al.*, 1994]. For Krakatau the Crête sulfate acidity is observed to rise abruptly in 1885 but then to decrease monotonically until it is lost in the background after 1887.

Based on this circumstantial evidence, the Montpellier pyrheliometric observations can be adjusted in the following way. Assuming the law of large numbers, the 3-year (1884–1886) average of the monthly turbidities is accepted to be a reliable number; this average turbidity is 0.07. According to the Crête ice core data the e-folding time for decay of the dust cloud was about 1 year. Consequently, the corrected annual turbidities for 1884, 1885, and 1886 are 0.14, 0.05, and 0.02.

The adjusted 1884 average turbidity can be checked by *Cornu*'s [1885] visual observations of the polarization of blue sky light over France. The monthly maximum percentage of polarization dropped in 1884 to 48% (determined also by *Pickering* [1886] in Massachusetts) from a previous, normal maximum of 75%. This corresponds to a decline of the monthly mean percentage of polarization from 58 to 37%, using *Kimball*'s [1913a] data to convert approximately from monthly maximum percentage to monthly mean percentage. According to Figure 1 the 1884 polarization decline would have been generated by a pyrheliometric turbidity of roughly 0.10. Averaging this result with the adjusted Montpellier turbidity of 0.14 gives a best overall turbidity estimate of 0.12 for 1884.

An average turbidity of ~0.1 is consistent with the reported intermittency of colored twilights throughout 1884. By the summer of 1885 the polarization decline had moderated; the monthly maximum percentage of polarization, as measured from Massachusetts, was 60% [*Pickering*, 1886]. This implies a pyrheliometric turbidity of about 0.04, which is in essential agreement with the adjusted Montpellier value of 0.05.

Another piece of information comes from a rough calculation by *Deirmendjian* [1973]. He utilized the fact that a sunspot had been seen with the naked eye on the dim green solar disk within half an hour before sunset on September 11, 1883, at Madras, India (13°N) [*Symons*, 1888; *Michie Smith*, 1883]. Assuming that the Sun was elevated 3° above the horizon, he derived an excess visual turbidity of 0.63. This corresponds to an excess pyrheliometric turbidity of approximately 0.40 if it was all due to small aerosols. Since the lapsed time from the Krakatau eruption was only 16 days, and a major buildup of sulfate aerosols may require several weeks to complete, a considerable amount of this excess turbidity could have been due to suspended ash particles. Furthermore, the dust cloud at this time was confined almost entirely to the tropical band. Therefore Deirmendjian's interesting result can only be used with caution to infer the stratospheric aerosol conditions in late 1883.

Astronomical reports shed very little further light on the question. Published data on stellar visibilities at night and in the daytime indicate that the astronomical extinction was perceived to be about 1–2 mag during the period between January 1884 and the summer of 1885 [*Fellow of the Royal Astronomical Society (FRAS)*, 1884; *Krone*, 1884; *Hazen*, 1884; *Winlock*, 1884; *Leeming*, 1884; *Hooper*, 1884, 1885]. These visual estimates can be only upper bounds on the true extinction, in view of the unknown zenith angles of the stars and the poor contrast between the "blurred" star images [*Symons*, 1888, p. 225] and the hazy sky background. Another limitation is that eye estimates of stellar magnitudes at that time had uncertainties of typically half a magnitude, even under favorable circumstances [*Gore*, 1884].

4.2. Tarawera 1886

The volcano Tarawera (38°S) in New Zealand exploded with enormous force on June 10, 1886. However, the volume of debris ejected was an order of magnitude smaller than Krakatau's, and the top of the eruption column came up well short of the tropopause [*Walker et al.*, 1984]. Not surprisingly, southern hemisphere observers noted no apparent optical disturbances in the upper atmosphere [*Fraser*, 1887].

In the two subsequent quiet periods, 1887–1889 and 1895–1901, there were again no large stratosphere-perturbing volcanic eruptions, at least not in the northern hemisphere. The lack of notable purple twilight glows and, especially, the absence of new sky polarization anomalies during these two periods [*Busch*, 1903] constitute sufficient proof.

4.3. Unidentified Eruptions of 1890 and 1893

Between August 1890 and January of the following year a number of magnificent purple twilight glows were reported across Germany [*Volger*, 1890; *Anonymous*, 1891]. These reports also included a sighting of Bishop's ring in December.

From Arnsberg in Germany, *Busch* [1891] observed that at some undetermined date between May 1890 and February 1891 the Arago and Babinet neutral points of sky polarization shifted dramatically, as they had after the Krakatau eruption. As time went on, the neutral points began to return to their normal positions, but in the early months of 1893, another abrupt displacement occurred. The disturbances finally disappeared in April 1895 [*Busch*, 1896].

The atmospheric disturbances seen in 1890–1891 can be detected in other data. Most observers of the total lunar eclipse of May 23, 1891, describe it as having displayed the usual ruddy tint across the Moon's surface. This illumination, however, was noted also as being rather faint and incompletely extended over the whole disk [*Booker*, 1891; *Lewitzky*, 1891; *Vérêchaguine*, 1891; *Wooster*, 1891], signifying the presence of some extinction in the Earth's atmosphere. The next total lunar eclipse, on November 15 of the same year, was widely reported on and appeared entirely normal, as if the extinction had by then decayed to an insignificant level.

Pyrheliometric measurements made at Montpellier revealed a small optical depth perturbation running between late 1890 and early 1892, as *Kimball* [1924] first noted. *Savelief* [1891], who was observing at Kiev, did not notice any decrease of sky transmission during this period, but his measurements are very sparse and certainly not systematic. The average turbidity inferred from the Montpellier data is 0.03, 0.06, and 0.03 for 1890, 1891, and 1892 (Figure 3).

Whatever eruption caused the later polarization anomalies of 1893–1894 left no trace in other kinds of turbidity records. Pyrheliometers at Montpellier and Pavlovsk showed no apparent signal.

The erupting volcanoes that were responsible for the small atmospheric disturbances of 1890 and 1893 have never been definitely identified. Sometimes suggested are Bogoslof (54°N) in the Aleutian Islands, which erupted in February 1890, and Awu (4°N) on Sangihe Island, Indonesia, which did the same in June 1892. However, our best estimates for the dates of the mysterious injections of stratospheric dust are June–August 1890 and January–March 1893.

4.4. Soufrière, Pelée, and Santa Maria 1902

Two volcanoes in the West Indies underwent a series of eruptions during the first half of May 1902. The biggest of the convulsions occurred on May 7 inside Soufrière of St. Vincent (13°N) and on May 8 inside Pelée of Martinique (15°N). Later in the year on October 24 and 25, Santa Maria (15°N) in Guatemala erupted in a great paroxysm that dwarfed the two West Indian eruptions. Nevertheless, Pelée has always drawn the most public attention because it destroyed the populous town of St. Pierre.

Immediately following the May 7–8 events, highly colored sunsets and twilight afterglows were seen over the Caribbean Sea [*Kimball*, 1902]. They appeared over Honolulu 12 days later [*Bishop*, 1902] and over Madeira Island in at most another 18 days [*Krohn*, 1902]. Since a careful pyrheliometric observer in the Pyrenees [*Marchand*, 1905] reported a significant diminution of sunlight as early as May 27, the westward velocity of the dust cloud must have been 13–18 m s^{-1}. The purple twilight glows continued, in subdued form, for 6 months [*Heilprin*, 1908]. Observed nightly durations of the glows indicated that the dust in June and October was concentrated at heights over Europe of 8–40 km [*Herschel*, 1902; *Marchand*, 1905] and 7–25 km [*Esclangon*, 1903], respectively, suggesting a drop in mean altitude from ~24 to ~16 km. At the end of October the glows suddenly intensified [*Forel*, 1905]. This rejuvenation, which was caused by the eruption of Santa Maria, lasted until February 1903, whereupon the glows weakened again until the following August, then renewed temporarily, and finally died out in 1904 [*Láska*, 1904; *Gruner*, 1914].

Bishop's rings appeared sporadically in many parts of Europe from late July to October 1902 [*Heilprin*, 1908]. Beginning

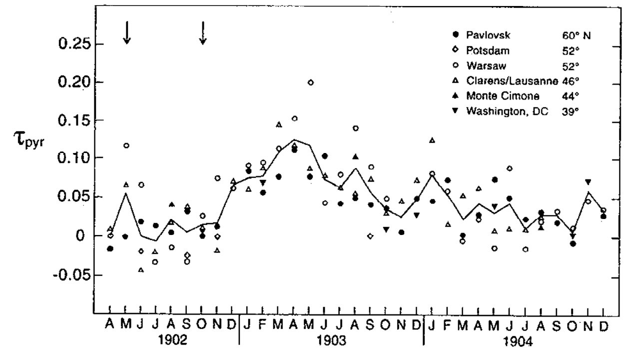

Figure 4. Pyrheliometric optical depth perturbations due to the eruptions of Soufrière and Pelée (May 1902) and of Santa Maria (October 1902). A mean curve is also shown. Arrows indicate the dates of the eruptions.

in November, such sightings became almost continual and only ceased sometime in 1904 [*Forel*, 1905].

At the same time, the Babinet and Arago neutral points of sky polarization showed a noticeable displacement from their normal positions, also beginning in November 1902 [*Busch*, 1903]. They were furthest displaced in April 1903 [*Sack*, 1904; *Busch*, 1905]. The percentage of blue sky light that was polarized, however, reached a minimum slightly later, in June of that year [*Kimball*, 1910]. As after Krakatau, the neutral points required about 4 years to completely return to normal [*Jensen*, 1937].

Pyrheliometric optical depth perturbations are displayed for this period in Figure 4. A sharp pulse of high turbidity appeared over Europe in late May 1902, following soon after the two West Indian eruptions. This volcanic signal, however, was absent or muted at the most northerly station, Pavlovsk (60°N), and seems to have been undetected in the northeastern United States [*Davis*, 1903; *Abbot*, 1903], probably owing to patchiness of the dust cloud. From June to October the turbidity over Europe was not very noticeable. Astronomical transparency of the sky, though somewhat poor [*Wolf*, 1903; *Barnard*, 1903], deteriorated in an obvious way only after November [*Backhouse*, 1903].

A stronger manifestation of turbidity occurred just after Santa Maria's eruption. The optical depth perturbation at northern midlatitudes reached a maximum of 0.12 in April 1903 and, subsequently, started a slow exponential decline, disappearing into the background by the beginning of 1905. Detection of this turbidity in the northeastern United States [*Kimball*, 1903a, b; *Abbot and Fowle*, 1908] testifies to the much greater eruptive magnitude of Santa Maria as compared to Soufrière and Pelée.

Although no useful pyrheliometric data from the southern hemisphere are available, two very dark lunar eclipses occurred in 1903 [*de Vaucouleurs*, 1944]. Their darkness strongly suggests that the volcanic cloud spread far south of the equator. This conclusion is reinforced by the roughly equal rates of aerosol fallout onto the Greenland and Antarctic ice sheets, as deduced from modern measurements of sulfate acidity in polar ice cores [*Hammer et al.*, 1980; *Legrand and Delmas*, 1987].

4.5. Ksudach 1907

The remote volcano Ksudach (52°N) on the Kamchatka peninsula of Russia erupted violently on March 28, 1907.

Unaware of this great eruption, sky observers in Germany and Switzerland began to notice moderate atmospheric anomalies in late April 1907. These anomalies strengthened in mid-May and then dwindled off and on until February 1909. They consisted of purple twilight glows, Bishop's rings, and high noctilucent clouds [*Wolf*, 1908; *Busch*, 1910; *Gruner*, 1914]. Accompanying the visible phenomena were small displacements of Babinet's and Arago's neutral points of sky polarization [*Busch*, 1908]. Observations at Tortosa, Spain [*Jensen*, 1928] and near Washington, D. C. [*Kimball*, 1913a], confirm that the percentage of polarization of blue sky light also fell slightly in 1907, although it recovered the next year. Contemporary observers recognized the clear analogies with Krakatau and Pelée, but they mistakenly attributed the atmospheric disturbances to the relatively minor outbursts of some of Italy's active volcanoes.

Pyrheliometric measurements made at Washington, D. C., [*Kimball*, 1910] showed a sudden drop in solar radiation on April 3, 1907, implying a southeastward movement of the dust cloud at 17 m s^{-1}. On the other hand, *Wolf*'s [1908] detection of the first twilight glow at Heidelberg, Germany, on April 28 suggests an average eastward velocity of only 5 m s^{-1}. The discrepancy might be resolved by any of the following possibilities: two layers of dust, vertical shearing, or patchiness, with rapid meridional movement.

In Figure 5 the available pyrheliometric optical depth perturbations are plotted for 1907–1909. At latitudes north of 35°N the turbidity started to increase in April 1907, reached a maximum value of 0.07 in May, then subsided into the background after March 1908. *Wolf* [1908] at Heidelberg also noticed a moderate reduction of the astronomical sky transparency in May and June 1907.

At Simla (31°N) in India, however, the measured changes of turbidity were insignificant. This negative result is supported by the long series of black and bright bulb actinometer measurements at Helwan (30°N) in Egypt [*Curry*, 1913]. It is almost certain therefore that the atmosphere was essentially undisturbed at latitudes south of 35°N. The total eclipse of the Moon on July 25, 1907, appeared perfectly normal [*de Vaucouleurs*, 1944].

4.6. Katmai 1912

On June 6, 1912, two sister volcanoes in Alaska, Katmai and Novarupta (58°N), began several days of violent eruption. Novarupta's massive eruption column successfully pierced the tropopause but Katmai's smaller one did not. Nonetheless, the series of events is usually called the "Katmai eruption."

From published twilight observations and pyrheliometer measurements *Volz* [1975a] has reconstructed in detail the time development of the dust cloud. The dust particles at first flowed southeastward at an average velocity of 17 m s^{-1}. By late June the dust had reached southern California, the eastern United States, Algeria, and Europe. Thereafter, purple twilight glows temporarily disappeared. After a sudden recommencement in early October, they persisted sporadically into late 1913 and possibly to the end of 1914. The bulk of the dust was located at altitudes above 15 km.

Bishop's rings and significant shifts of the Arago and Babinet neutral points first appeared, as expected, in late June 1912 [*Kimball*, 1913a] and continued well into 1914 [*Maurer and Dorno*, 1914; *Jensen*, 1928, 1937]. The percentage of blue sky polarization fell to a minimum around the time of greatest turbidity, in August 1912 [*Kimball*, 1913a].

Pyrheliometric optical depth perturbations, derived by *Volz* [1975a], have been recalculated here for the following reasons.

First, Volz chose to use only days with the most transparent skies, whenever possible, whereas we have preferred to use all clear days in each month. Second, a few of the stations used by Volz possess pyrheliometric records that cannot be adequately calibrated in our view, and so these stations have been omitted here. Third, the extensive data from the *Madrid Observatory* [1911–1920] have been added. Finally, it seems desirable to present a turbidity record for Katmai that is completely consistent with those for the other eruptions. In the event, our latitudinally averaged monthly mean turbidities for Katmai turn out to be almost identical to Volz's results.

The individual monthly mean turbidities are plotted against time in Figure 6. For latitudes between 45°N and 60°N they display a flat maximum, averaging 0.23 during the summer of 1912. Then they abate more or less exponentially, reaching a negligible level by October 1914. Their development is very similar at latitudes between 30°N and 45°N, except that their maximum value there is only 0.13. Station-to-station variations within these latitude bands obviously exist, but the formally derived variations cannot be trusted as being physically real. The noise level, which is determined by various sources of accidental error as well as by real scatter, lies at about ±0.03, as judged from Figures 4–6.

No significant excess turbidity was detected during the years 1912–1914 at Mexico City (19°N) [*Galindo and Muhlia*, 1970] or at Arequipa, Peru (16°S) [*Abbot*, 1916]. Slight apparent variations of turbidity over Arequipa in those years probably had local causes [*Volz*, 1975a]. After January 20, 1913, a minor increase of turbidity appeared briefly over Mexico City, but it was probably due to the recent eruption of Colima only 300 km away.

Observations of anomalous stellar extinction were also made during the summer of 1912. Visual measurements of the near-zenith extinction over Europe tended to be high: 1–3 mag [*Alker*, 1913; *de Roy*, 1913; *Wolf*, 1913]. Most of the United States measurements gave somewhat lower values [*Kimball*, 1913b]. All these eye estimates can probably be discounted just as in the case of the 1884–1885 observations. On the other hand, a reasonably good photographic (λ = 0.425 μm) determination of the stellar extinction over Minnesota by F. P. Leavenworth [*Kimball*, 1913b] yielded 0.5–1.0 mag; this value converts into a pyrheliometric turbidity of 0.2–0.4, close to what pyrheliometers measured at that time.

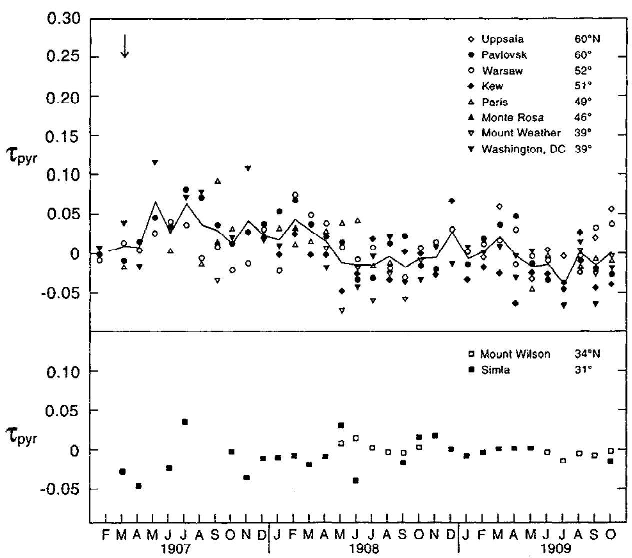

Figure 5. Pyrheliometric optical depth perturbations due to the eruption of Ksudach (March 1907). A mean curve is also shown. The arrow indicates the date of the eruption.

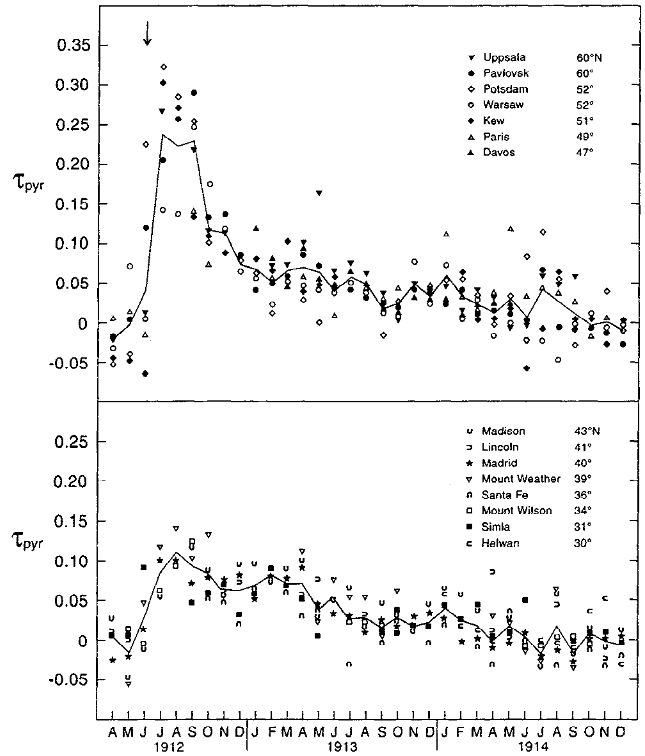

Figure 6. Pyrheliometric optical depth perturbations due to the eruption of Katmai (June 1912). A mean curve is also shown. The arrow indicates the date of the eruption.

No appreciable extinction of starlight was observed at Helwan, Egypt (30°N) (H. Knox-Shaw in the work of *Curry* [1913]) or at Santiago, Chile (33°S) (J. H. Moore in the work of *Knoche* [1913]). These negative results confirm that the aerosol veil was effectively absent from latitudes south of 30°N.

A total eclipse of the Moon on March 22, 1913, was very dark [*de Vaucouleurs*, 1944], indicating that the Moon passed through the Earth's umbra in the section that was cast by the Earth's northern latitudes.

4.7. Puyehue 1921

Puyehue Volcano (41°S) in central Chile erupted on December 13, 1921, sending up a dust cloud that swept over La Plata in Argentina four days later and, apparently, Windhoek in Namibia 10 days after that [*Wolf*, 1924]. The average eastward velocity was 3–7 m s^{-1}.

In northern Chile the Mount Montezuma (23°S) APO station recorded a small but significant drop in solar radiation during the year following the eruption. Thus in 1922 the optical depth perturbation averaged 0.011, according to the final data published by *Abbot et al.* [1932]. Although these data are very close to those that *Abbot et al.* [1923] first published, they differ from the tentatively revised data of an intermediate date [*Abbot*, 1926], which indicated an average optical depth perturbation of about zero. Because *Abbot* [1926] expressed much doubt about the reliability of both the observers and the instrumental apparatus in 1921 and 1922, we do not regard the Mount Montezuma measurements as being very secure for this

Table 5. Monthly Mean Pyrheliometric Turbidities After the Eruptions of Paluweh (August 1928) and Quizapu (April 1932)

	Mount Brukkaros		Mount Stromlo	
Month	1928	1929	1932	1933
January		0.011		0.012
February		−0.007		0.002
March		0.018	−0.004	−0.005
April		0.005	0.018	
May		0.007	0.026	
June		0.001	0.014	
July		−0.003	0.025	
August	−0.001		0.030	
September	−0.005		0.020	
October	0.000		0.032	
November	−0.001		0.017	
December	−0.004		0.002	

Data are from *Abbott et al.* [1932] for Mount Brukkaros and from *Rimmer* [1937] for Mount Stromlo. For Mount Montezuma data, see *Hoyt*'s [1979a] Table 5.

period. Nevertheless, the 1932 version of the data is accepted here, and the resulting optical depth perturbations are listed in Table 6.

Northern hemisphere consequences of the eruption were apparently negligible, as judged from the series of pyrheliometric measurements made at Davos [*Lindholm*, 1927] and at Pavlovsk [*Kalitin*, 1926]. *Kimball*'s [1924] plotted monthly mean transmissions for four United States stations, Lincoln, Madison, Santa Fe, and Washington, D. C., also indicate no significant perturbation. This result casts serious doubt on the claims of *Wolf* [1924] and *Dorno* [1925] that the occasional atmospheric anomalies that were seen in the northern hemisphere during 1922 arose from the Chilean dust cloud. Sporadic anomalies of the type and frequency they reported can and do arise from nonvolcanic causes in any given year. Moreover, it is highly unlikely that any significant amount of dust from such a southerly eruption would have crossed the equator.

Between October 1920 and March 1921 a slight depression of sky transmission occurred in the northern hemisphere, according to *Kimball* [1924]. The improved reduction procedures used here do not confirm this depression, and it certainly lies well within the noise level of Kimball's displayed plot. A comparably small drop that Kimball detected in the measurements of *Abbot et al.* [1923] was probably caused by the relocation of the APO measuring apparatus from Mount Wilson, California, to Mount Harqua Hala, Arizona, in September 1920. The Mount Harqua Hala measurements continued to deteriorate in quality until at least late in 1922 [*Abbot et al.*, 1923].

4.8. Paluweh 1928 and Komagatake 1929

During August and September 1928, a series of eruptions shook the Indonesian island volcano of Paluweh (8°S), just north of Flores. The expelled dust is believed to have caused small decreases in worldwide atmospheric transmission during the following 14 months [*Hand*, 1939; *Hoyt*, 1978, 1979a].

Hoyt [1979a] demonstrated from APO data that the average optical depth perturbation between October 1928 and March 1929 was 0.013 at Mount Montezuma (23°S). However, the perturbation at Mount Brukkaros (26°S), Namibia, averaged only 0.003 (Table 5) if we use the simultaneously obtained APO data for that site [*Abbot et al.*, 1932]. Owing to the reported frequency of desert dust blowing over Mount Brukkaros, the Mount Montezuma results are probably more reliable and are therefore adopted here.

In the northern hemisphere, *Hand* [1939] detected small, though ill-defined, diminutions of atmospheric transmission between March and November 1929 at Lincoln, Madison, and Washington, D. C. During approximately the same period at the clearer site of Table Mountain (34°N), California, the APO observations apparently confirmed these decreases [*Hoyt*, 1979a], as did the Davos (Switzerland) observations [*Hoyt and Fröhlich*, 1983].

To sum up, the available data suggest an average turbidity increase of roughly 0.01 in late 1928 in the southern hemisphere, with a smaller increase coming five months later in the northern hemisphere. If Krakatau can serve as a guide, an alternative interpretation might be that the dust veil was restricted to the tropics in both hemispheres before March 1929, and then rapidly expanded poleward.

At any rate, it is meaningless to average the monthly APO observations from Mount Montezuma, Mount Brukkaros, and Table Mountain to form a composite time curve. Although *Abbot et al.* [1932] did so, this was before the potential importance of the Paluweh eruption was recognized. Unfortunately, their Figure 26b conveys a false impression (which misled *Hoyt* [1979a, b]) about the relative strengths of the Paluweh and Puyehue dust veils, Paluweh's seeming to be much weaker.

Self et al. [1981] have questioned whether the Paluweh eruption was large enough to have produced the optical depth perturbations of late 1928 and early 1929. The brief descriptions of the eruption given by *Neumann van Padang* [1951] and *Hand* [1939] suggest that it was [*Hoyt*, 1978], and the timing coincidence is strong.

On the other hand, an abrupt rise of the turbidity to a value of 0.03 at Table Mountain in July and August 1929 is difficult to attribute to Paluweh's dust veil. It was probably caused by the large eruption of Komagatake (42°N) in Japan on June 17, which Self et al. also mentioned. The negligible turbidities detected over Mount Montezuma between mid 1929 and the end of 1931 indicate that no significant amount of dust from this northern eruption drifted into the southern hemisphere.

4.9. Quizapu 1932

Another Chilean volcano, Quizapu, the main crater of Cerro Azul (36°S), erupted on April 10, 1932. This eruption was of comparable size to Puyehue's in 1921. Within 2 weeks, brilliant twilight glows appeared over South Africa [*Blathwayt*, 1932]. Since the east-moving dust subsequently reached New Zealand on or about May 5 [*Kidson and Simmers*, 1933], it must have traveled at an average speed of about 9 m s^{-1}.

To the north the Mount Montezuma (23°S) APO station registered a mean optical depth increase of 0.021 over the next six months [*Hoyt*, 1979a]. At Mount Stromlo (35°S) in Australia [*Rimmer*, 1937] the average increase amounted to 0.024 during the same period (Table 5).

It is very improbable that the northern hemisphere received any significant dust from this Chilean eruption. Pyrheliometers located in Lincoln, Madison, and Washington, D. C. [*Hand*, 1939] recorded no perceptible diminution of solar radiation during 1932 and 1933. Fluctuations seen at Table Mountain [*Aldrich*, 1945; *Hoyt*, 1979a] and at Davos [*Hoyt and Fröhlich*, 1983] lie within the noise levels for these two stations.

4.10. Spurr 1953

When Mount Spurr (61°N) in Alaska erupted on July 9, 1953, thin stratospheric dust clouds were observed 2–3 weeks later over the British Isles [*Jacobs*, 1954] and over Germany [*Volz*, 1954]. A total eclipse of the Moon on July 26 appeared to be abnormally dark [*de Vaucouleurs*, 1954], and purple twilight glows were occasionally reported from central Europe during August and September [*Volz*, 1954]. It seemed that a major volcanic perturbation to the stratosphere was under way.

Pyrheliometric data for the 6 months following the July eruption, however, indicated otherwise. In the United States, only very minor changes in atmospheric transmission (conflicting among themselves in sign) were recorded at Albuquerque, Blue Hill (Massachusetts), Lincoln, Madison, and Table Mountain [*Fritz*, 1956; *Hoyt*, 1979a, b; *Hoyt et al.*, 1980]. All of these fluctuations remained within the long-term noise levels for these stations. Similarly, nothing significant was recorded at Davos [*Hoyt and Fröhlich*, 1983].

In late 1952, just a few months before Spurr erupted, a small drop in atmospheric transmission occurred at Mount Montezuma and lasted about 2 years [*Hoyt*, 1979a, b; *Roosen and Angione*, 1984]. The cause of the drop remains unknown, although local pollution and an unidentified southern hemisphere eruption have been suggested.

4.11. Bezymianny 1956

The enormous eruption of Bezymianny (56°N) in Kamchatka, Russia, on March 30, 1956, produced only a small stratospheric dust cloud, much like Spurr's three years before. The cloud was detected over the British Isles on April 3 [*Bull and James*, 1956].

Pyrheliometric observations made at Davos [*Hoyt and Fröhlich*, 1983], at Albuquerque, Blue Hill, and Madison [*Hoyt et al.*, 1980], and at Tucson [*Heidel*, 1972] show no significant stratospheric disturbance in 1956.

5. Chronology of Growth and Decay of Dust Clouds

From the pyrheliometric data assembled in the preceding sections, a chronology of the monthly mean optical depth perturbations has been constructed. The numerical results are entered as a function of latitude band in Table 6, where blank and missing entries should be read as zero. Note that these are pyrheliometric turbidities; to obtain visual turbidities, multiply all entries by 1.6 (bearing in mind the caveats of section 2.2). Since the table contains unfiltered data, the user can smooth these data in space and time according to the user's own requirements. (The table is not available in electronic form.)

Some remarks about the construction of the table will be helpful. After the first few months in the two earliest volcanic periods (the first one in a section running from January 1884 to December 1886 and the other in a section from December 1890 to December 1892) the optical depth perturbation was assumed to decay exponentially with time, the e-folding time being set equal to 1 year. In the absence of other information the highest section of the rising part of the curve, occurring just before the turbidity peak, was simply made to be symmetrical with respect to the decaying part. Judging from the detailed data available for several later eruptions, these two assumptions should be not too far off the mark.

In the case of the first volcanic period, some information is available about the optical depth perturbation in September 1883, which has been used to roughly estimate turbidities between then and the end of the year. The 1890–1892 curve has an arbitrary latitude range (0° to 90°N) assigned to it, because the volcano responsible for the dust veil is unknown and because observations were reported only from 43°N to 51°N.

The period between November 1902 and the early months of 1903 is confused by the effects of three eruptions. The combined Soufrière-Pelée dust veil, which originated in May 1902, is assumed to have continued to decay during this period of overlap with an e-folding time of 1 year. Santa Maria's dust cloud, thrown up in late October, is allowed to suddenly become global in February 1903, since all that Figure 4 and other available evidence imply is a date somewhere between December 1902 and April 1903.

Tabulated latitude ranges for Puyehue's and Quizapu's dust veils (in both cases 0° to 90°S) are somewhat uncertain, because published observations of these dust veils were reported only from latitudes between 22°S and 41°S. It is certain, however, that no significant amount of dust from the eruptions went into the northern hemisphere.

For all these eruptions no useful pyrheliometric data are available from polar and subpolar observing stations. Owing to the small height of the tropopause at high latitudes, stratospheric aerosol particles that have been transported poleward from lower latitudes settle out more slowly than they do at midlatitudes and so reach a maximum accumulation above the poles roughly a year after a large low-latitude eruption. This circumstance is well known from a variety of studies of atmospheric nuclear test explosions [*Volz*, 1975b], volcanic acidities in Greenland ice cores [*Hammer et al.*, 1980; *Fiacco et al.*, 1994], and stratospheric dust veils due to recent tropical eruptions [*Sato et al.*, 1993; *Trepte et al.*, 1994]. Nevertheless, in the absence of specific information the monthly turbidities for the older eruptions are assigned here to have the same values at high latitudes as at midlatitudes.

After a volcanic dust veil reaches its state of maximum turbidity, the turbidity decays more or less exponentially. Sufficiently long records are available for the Santa Maria and Katmai dust veils to estimate approximate e-folding times by the method of least squares. We thus obtain

$$t_0 = 0.9 \pm 0.2 \text{ year} \qquad \text{(Santa Maria)}$$

$$t_0 = 0.8 \pm 0.1 \text{ year} \qquad \text{(Katmai).}$$

Volz [1975b] likewise inferred $t_0 = 0.8$–1.0 year for Katmai. These results are not statistically different from the standard value of about 1 year estimated for other large eruptions, such as Tambora (1815) [*Stothers*, 1984a], Agung (1963) [*Volz*, 1970], and Fuego (1974), El Chichón (1982), and Pinatubo (1991) [*Rosen et al.*, 1994].

No attempt has been made here to solve for a possible annual cycle for the meridional dust transport. The available data for the pre-Agung eruptions are inadequate.

Mean annual optical depth perturbations for each hemisphere have been prepared from the data in Table 6 and are plotted as visual means in Figure 7. These results are expected to be an improvement over the provisional values published by *Sato et al.* [1993] for the corresponding years. (The values of Sato et al. should be multiplied by $k = 1.6$ in order to be compared with ours.) For the more recent period the Sato et al. unmodified values form the extension shown in Figure 7,

Table 6. Pyrheliometric Optical Depth Perturbations (Multiplied by 1000) Due to Volcanic Eruptions During the Period 1881–1960

Year	Lat Range	Jan.	Feb.	March	April	May	June	July	Aug.	Sept.	Oct.	Nov.	Dec.
1883	−90° to −30°												168
	−30° to +30°									400	370	340	168
	+30° to +90°												168
1884	−90° to +90°	182	168	154	142	130	120	110	102	93	86	79	73
1885	−90° to +90°	67	62	57	52	48	44	41	37	34	32	29	27
1886	−90° to +90°	25	23	21	19	18	16	15	14	13	12	11	10
1890	0° to +90°								40	77	84	91	99
1891	0° to +90°	91	84	77	71	65	60	55	51	47	43	40	36
1892	0° to +90°	33	31	28	26	24	22	20	19	17	16	15	13
1902	−90° to −30°						0	0	20	6	14	11	10
	−30° to +90°						0	0	20	6	14	17	67
1903	−90° to −30°	9	77	110	125	119	74	63	89	59	37	27	49
	−30° to +90°	76	77	110	125	119	74	63	89	59	37	27	49
1904	−90° to +90°	83	53	24	45	32	45	14	30	29	9	61	34
1905	−90° to +90°	17	12	21	31	0	12						
1907	+30° to +90°				8	67	27	64	37	30	12	43	26
1908	+30° to +90°	19	44	32	17								
1912	+30° to +45°						29	86	112	95	84	66	63
	+45° to +90°						40	237	223	228	118	115	76
1913	+30° to +45°	70	83	75	74	39	55	30	31	19	33	23	26
	+45° to +90°	69	52	65	71	65	44	58	49	18	23	50	37
1914	+30° to +45°	43	28	21	3	20	9	0	21	0			
	+45° to +90°	60	33	25	14	31	8	46	27	12			
1922	−90° to 0°	4	0	2	10	12	13	19	11	16	15	13	11
1923	−90° to 0°	13	11	8	9	10	8	7					
1928	−90° to 0°										11	17	7
1929	−90° to 0°	6	25	14	7	14	10	12	2	6	2		
	0° to +90°			2	1	7	2	34	28	8	18	7	8
1932	−90° to 0°				11	24	21	21	25	15	29	20	7
1933	−90° to 0°	9	10	2	13								

Blank and missing entries are to be taken as zero.

including an update to 1994 (M. Sato and J. Hansen, personal communication, 1995).

The Krakatau period as reconfigured reveals one large change from the earlier reconstruction: the optical depth maximum has shifted from 1885 to 1884. For Santa Maria the two hemispheres now show equal turbidity. The revised optical depth maxima for Ksudach and Katmai are about half the values estimated by Sato et al. Noise in the earlier record has been identified and eliminated. Puyehue did not previously appear. Although the other differences are small, our new values rest on a more secure basis, especially at the monthly level.

Unmistakable in the trends shown by Figure 7 is a long quiet period that extends between 1913 and 1962. This is due to a lack of very large, sulfur-rich volcanic eruptions during these years and corresponds to a drop in the number of eruptions of all types large enough (VEI $\geq$ 4) to have left an essentially homogeneous record of volcanism [*Newhall and Self*, 1982]. Further discussion of this point is deferred to section 8.

6. Masses of the Dust Veils

Global mass loadings of the stratosphere can be calculated from the derived aerosol optical thicknesses. Adopting spherical coordinates and assuming azimuthal symmetry of the dust distribution, the total aerosol mass (see Appendix) is

$$M_D = 2\pi R^2 \mu \int_{-\pi/2}^{\pi/2} \tau_D \cos\phi \, d\phi \qquad (9)$$

where R is the radius of the Earth (neglecting the height of the aerosol layer above the surface), ϕ is the geographical latitude, and μ is an effective aerosol column mass per unit optical depth. We adopt for aerosol particle parameters: $Q\lambda/(2\pi r) = 0.64$, valid for a sulfate refractive index of 1.4–1.5 in the visual band [*van de Hulst*, 1957; *Lacis and Mishchenko*, 1995], and $\rho = 1.65$ g cm^{-3}. Therefore $\mu(\lambda 0.55) = 3.0 \times 10^{-5}$ g cm^{-2}. Hence, in units of megatons (Tg),

$$M_D = 75 \int_{-\pi/2}^{\pi/2} \tau_{\text{vis}} \cos\phi \, d\phi. \qquad (10)$$

This formula is rather insensitive to the sizes and hence size distribution of the particles if their effective radius, r_e, lies in the range 0.15–0.5 μm. The expected error of μ is less than 20%.

Calculation of the total aerosol mass must be based on the amount of turbidity observed during the month of peak turbidity. By this time, most of the erupted sulfur gases have converted to sulfate aerosols, but very little fallout of the aerosols from the stratosphere has occurred. Sulfur conversion appears to require only a few weeks to be essentially completed [*Read et al.*, 1993]. But since the aerosols are initially small and are located at very high altitudes, they probably do not begin to settle out in significant numbers for 3–6 months, judging both from the prolonged flatness of the observed turbidity maxima and from the direct observations of fallout from atmospheric nuclear test explosions [*Volz*, 1975b]. In the case of tropical eruptions, because of the circumstance that the available pyrheliometer stations are not located in the original latitude

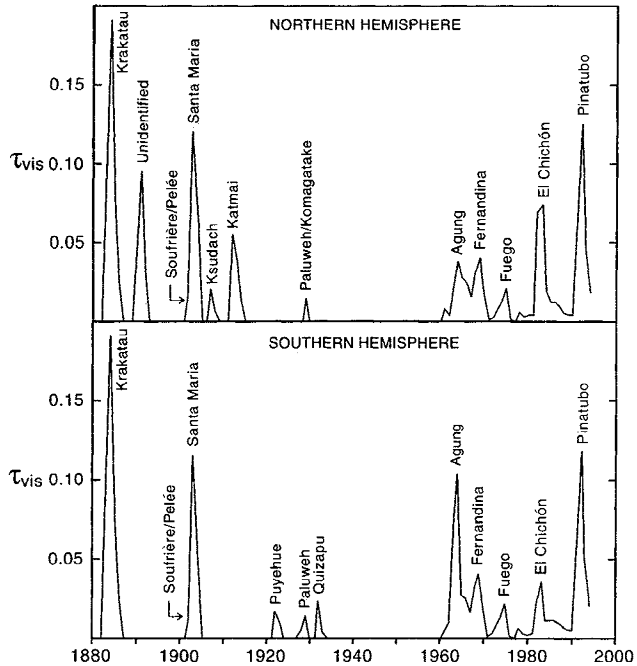

Figure 7. Mean annual optical depth perturbations at $\lambda = 0.55\ \mu m$ for the northern and southern hemispheres. Data for the most recent years, 1961–1994, come from *Sato et al.* [1993] and Sato and Hansen (personal communication, 1995). Maxima are labeled with the source volcano.

band of the injected dust veil, it requires several months before the dust becomes fairly smoothly distributed over these stations, and therefore the turbidity there appears to peak late. Assuming that little fallout of the aerosols has yet taken place, we adopt the first month of the broad turbidity maximum as the measurement month in order to calculate the amount of volcanic aerosols that have formed. Use of an exponential extrapolation of the turbidity back to the time of the eruption [*Rosen et al.*, 1994] would probably lead to a considerable overestimate of the aerosol loading. The measurement month is identified in Table 7 for each volcanic eruption under present consideration. Integration over latitude of the τ_{pyr} profile for this month, with combined spectral and latitudinal weighting factors $k \cos \phi$, then yields the total aerosol mass listed in the last column of Table 7.

Derived mass values range from 2 Tg for the eruption of Puyehue (about the smallest mass that can be determined by this method under the best of circumstances) to 44 Tg for the eruption of Krakatau. Previous mass estimates for volcanic eruptions in this time period are listed in Table 8. The methods used include (1) a less accurate version of the present turbidity method, (2) the ice core method, (3) a petrologic method based on determining the sulfur content of glass inclusions in the erupted magma, and (4) a method that consists of simply assuming a constant sulfur percentage of the total ejected tephra mass. Method 3 usually yields an underestimate, while method 4 is crudely statistical; therefore these two methods will not be considered further here. The ice core method is more useful, although it must assume a latitude correction factor to allow for the stratospheric transport of aerosols from the eruption site to the site where the ice core was retrieved. All of these methods except the turbidity method yield a total mass of H_2SO_4 acids, not of H_2SO_4 aerosols. To convert between the two, we adopt a conventional model of the aerosols in which they consist of 75% H_2SO_4 and 25% H_2O [*Rosen*, 1971; *Toon and Pollack*, 1973].

Aerosol masses derived from the Greenland and Antarctic ice cores agree well with our turbidity-based values in the cases of Krakatau and Santa Maria. For Katmai, however, the Greenland ice core estimate [*Hammer et al.*, 1980] is 4 times as large as the turbidity-based value derived both here and by *Volz* [1975b]. The promptness of the aerosol fallout over Greenland in 1912 suggests a large direct tropospheric component. The fallout beginning a year after the eruption, ~10 Tg in amount, would have been stratospheric; this estimate agrees with our turbidity-based value. These agreements also confirm that $k \approx 1.6$ in the months of maximum turbidity.

7. Total Output of Sulfur Dioxide From Largest Eruptions

Throughout the period under present study, the average threshold for detection of a stratospheric dust veil is crossed when the aerosol mass loading equals approximately 4 Tg. After 1960, only three eruptions produced aerosol veils exceeding this limit: 20 ± 5 Tg from Agung (1963) [*Deirmendjian*, 1973; *Cadle et al.*, 1976; *Legrand and Delmas*, 1987], 14 ± 4 Tg from El Chichón (1982) [*McCormick and Swissler*, 1983; *Mroz et al.*, 1983; *Bluth et al.*, 1993; *Eparvier et al.*, 1994], and 30 ± 10 Tg from Pinatubo (1991) [*McCormick and Veiga*, 1992; *Bluth et al.*, 1993; *Baran and Foot*, 1994; *Rosen et al.*, 1994; *McPeters*, 1995]. All values for Agung have been adjusted to a loading ratio of 5:1 between the southern and northern hemispheres [*Volz*, 1970]. Although modern ground-based, airborne, and

Table 7. Volcanic Aerosol Loading of the Stratosphere, 1881–1960

Volcano	Lat	Long	Eruption Date	Measurement Date	M_D, Tg
Krakatau	6°S	105°E	August 26, 1883	January 1884	44
Unidentified			July (?), 1890	December 1890	6–24[a]
Soufrière/Pelée	14°N	61°W	May 7, 1902	August 1902	5
Santa Maria	15°N	92°W	October 24, 1902	April 1903	30
Ksudach	52°N	158°E	March 28, 1907	May 1907	4
Katmai	58°N	155°W	June 6, 1912	July 1912	11
Puyehue	41°S	72°W	December 13, 1921	1922 (average)	2
Paluweh	8°S	122°E	August 4, 1928	February 1929	3
Komagatake	42°N	141°E	June 17, 1929	July 1929	4
Quizapu	36°S	71°W	April 10, 1932	May 1932	3

[a]6 Tg, if a high-latitude eruption; 12 Tg, if a midlatitude eruption; 24 Tg, if a tropical eruption.

Table 8. Previous Determinations of Volcanic Aerosol Loading of the Stratosphere during the Period 1881–1960

Method	M_D, Tg			Source
	Krakatau	Santa Maria	Katmai	
Turbidity	48[a]		21	*Deirmendjian* [1973]
			13	*Volz* [1975b]
Greenland ice[b]	73	32[c]	40 [d]	*Hammer et al.* [1980]
Antarctic ice[b]	~45	29		*Legrand and Delmas* [1987]
	45			*Delmas et al.* [1992]
	64			*Langway et al.* [1988]
Glass inclusions[b]	≥4			*Devine et al.* [1984]
		≥8	≥11	*Palais and Sigurdsson* [1989]
Total erupted tephra	50	34	5	*Mitchell* [1970]

[a]Refers to a mixture of aerosols and ash particles two weeks after the eruption.
[b]Published mass loading of H_2SO_4 multiplied by 1.33.
[c]Deduced here from the Crête, Greenland, ice acidity record, using the method of *Hammer et al.* [1980].
[d]Should be multiplied by 1/4 (see text).

spacecraft instruments can now detect dust veils much less massive than 4 Tg, the purpose of using this limit is to be able to construct a fully homogeneous time series from 1881 to 1992.

The entire span of 112 years has been divided into eight short intervals of 14 years length. For each interval the summed mass of H_2SO_4 aerosols from volcanic eruptions that individually produced $M_D \geq 4$ Tg has been converted into a total mass of SO_2. The results are presented in Table 9.

During the most recent interval, 1979–1992, the mass of SO_2 from these large eruptions was 22 ± 5 Tg, in essential agreement with the Total Ozone Mapping Spectrometer satellite estimate of 30 ± 9 Tg [*Bluth et al.*, 1993]. However, the most recent interval is highly atypical of the whole period 1881–1992. Only the two earliest intervals, covering the years 1881–1908, were as productive. In the five intervening intervals the SO_2 output ranged from ~0 to 10 Tg. Over the entire 112-year period the average annual production rate was 0.8 Tg yr^{-1}, just half the average rate in the 1979–1992 interval. Any extrapolation to the future based only on recent years therefore must carry considerable uncertainty.

The output of SO_2 from a volcanic eruption is found to be only weakly correlated with the eruption's explosivity, as measured, for example, by its total volume of ejected tephra [*Rampino and Self*, 1984]. Following *Bluth et al.* [1993], we use as such an indicator of explosivity the volcanic explosivity index (VEI) of *Newhall and Self* [1982]. The four largest eruptions in the period 1881–1992 (Krakatau, Santa Maria, Katmai, and Pinatubo) had VEI = 6, yet they showed a range of a factor of 4 in their SO_2 outputs. Over the same period the seven VEI = 5 eruptions (Tarawera, Ksudach, Quizapu, Bezymianny, St. Helens, El Chichón, and Cerro Hudson) exhibited a range of a factor of 10 or larger. This wide variability in SO_2 output illustrates the potential risk of relying solely on the VEI (or the dvi) as a proxy for sulfate aerosol output, as is sometimes done.

Table 9. Sulfur Dioxide Loading of the Stratosphere by Volcanic Dust Veils With Aerosol Masses of at Least 4 Tg

Period	Mass of SO_2, Tg	Volcanic Eruptions (VEI)
1881–1894	28	Krakatau (6), unidentified (?)
1895–1908	19	Soufrière/Pelée (4), Santa Maria (6), Ksudach (5)
1909–1922	5	Katmai (6)
1923–1936	2	Komagatake (4)
1937–1950	0	
1951–1964	10	Agung (4)
1965–1978	0	
1979–1992	22	El Chichón (5), Pinatubo (6)

8. Summary and Conclusions

From this new study of stratospheric dust veils during the years 1881–1960 the principal results obtained are as follows:

1. Ten volcanic dust veils have been detected from pyrheliometric and other data. Time histories, derived in detail for the first time here, are given for the dust veils due to Krakatau (1883), Soufrière and Pelée (1902), Santa Maria (1902), Ksudach (1907), Puyehue (1921), and an unidentified eruption of 1890. Some improvements that have been obtained in the cases of Katmai (1912), Paluweh (1928), Komagatake (1929), and Quizapu (1932) are also presented.

2. Stratospheric optical depth perturbations for all of these eruptions are tabulated as a function of year, month, and latitude band. This homogeneous set of data is expected to be useful for climate modeling studies.

3. The visual turbidity exceeds the pyrheliometrically determined turbidity by a factor of 1.6 from about one month after the eruption until at least a few months later. This factor still has some uncertainty, however.

4. Dust veils, once built up in the lower stratosphere, decay approximately exponentially, as many authors have noted. The e-folding time was 0.9 years for Santa Maria and 0.8 years for Katmai.

5. A stratospheric dust veil is unlikely to be detected pyrheliometrically if $\tau_{pyr} < 0.03$.

6. Purple twilight glows following large eruptions are suppressed if $\tau_{pyr} > 0.09$.

7. The percentage of polarization of blue sky light declines with increasing turbidity in an approximately exponential manner.

8. Neutral points of blue sky polarization become more displaced as the turbidity increases. This long-known relation, however, is not wholly monotonic except at large Sun elevation angles.

9. Bishop's rings around the Sun show a remarkable uniformity of average size for all eruptions.

10. A spreading dust veil from a high-latitude eruption stays mostly poleward of 30°. One from a midlatitude eruption probably occupies the entire hemisphere in which it formed. If the

dust veil arises from a tropical eruption, it eventually spreads over the whole globe, although not necessarily equally in both hemispheres. These results confirm much earlier work.

11. Aerosol mass loadings of the stratosphere, derivable from the turbidities, agree well with published values based on polar ice core acidities, in the important cases of Krakatau, Santa Maria, and Katmai (after a major correction of the Katmai ice core value).

12. Volcanic production of stratospheric SO_2 follows from the aerosol mass loadings. Based on individual volcanic aerosol loadings of at least 4 Tg, the time-integrated SO_2 output was very high during 1881–1902, low during 1903–1981 (with a long 1913–1962 minimum), and high again during 1982–1992. Over the whole 112-year interval the average annual production rate of SO_2 from large eruptions was 0.8 Tg yr^{-1}, about half the rate during recent years.

13. Since 1881, the biggest single producer of stratospheric SO_2 was Krakatau, with Santa Maria and Pinatubo tied for second place.

Determination of the long-term trends in past times will ideally require specific information about aerosol production by the most explosive eruptions before 1881. Such early data can be best acquired either from visual turbidity estimates or from ice core acidity measurements. We have now shown that the two methods give highly concordant results in the prominent cases of the AD 536 eruption, Laki, Tambora, Krakatau, Santa Maria, and Katmai. In fact, a network of ice core measurements over the globe, together with some knowledge of the latitude of an eruption, can be used to invert equation (10) and to obtain a reliable estimate of the peak visual optical depth perturbation, as has been already explicitly demonstrated in the cases of the Tambora and AD 536 eruptions [*Stothers*, 1984a, b].

The Crête, Greenland, acidity record since AD 553 suggests that there exist long waves of volcanic activity [*Hammer et al.*, 1980], which the detailed record of global volcanism since 1500 likewise shows [*Simkin et al.*, 1981]. As these waves display a quasi-periodicity of ~80 years [*Stothers*, 1989], the remarkable lull in stratospheric aerosol production during the years 1913–1962 may not be atypical over the very long term. Such lulls will have to be factored into any calculations that attempt to reliably extrapolate future aerosol production from the present state of high volcanic activity.

Appendix

The total mass of volcanic aerosol particles suspended within the entire volume of the atmosphere at any given time is

$$M_D = \iiiint m(r)n(r)\, dr\, dx\, dy\, dz \qquad \text{(A1)}$$

where r is the characteristic particle radius, $m(r)$ is the particle mass, and $n(r)$ is the number of particles per unit volume per unit interval of r. The optical depth of a column of light scatterers in the vertical direction z at any horizontal position (x, y) is defined by

$$\tau_D = \iint \sigma(r)n(r)\, dr\, dz \qquad \text{(A2)}$$

where the light-scattering coefficient is

$$\sigma(r) = \pi r^2 Q \qquad \text{(A3)}$$

for spherical particles. The scattering efficiency factor, Q, depends on the refractive index of the particles and on the ratio $2\pi r/\lambda$, where λ is the wavelength of light in which τ_D is measured.

A unit column mass can be usefully defined in terms of σ and the particle mass density ρ:

$$\mu = m\sigma^{-1} = (4\pi r^3 \rho/3)\sigma^{-1}. \qquad \text{(A4)}$$

Thus if $Q\lambda/(2\pi r)$ is independent of r, so is μ. In that case,

$$M_D = \mu \iint \tau_D\, dx\, dy. \qquad \text{(A5)}$$

This result generalizes (and hence justifies) an expression derived previously for the restricted case of monodisperse particles that are uniformly distributed in space [*Stothers*, 1984a]. Its validity depends only on the assumption of the constancy of the two quantities ρ and $Q\lambda/(2\pi r)$, a condition that is approximately true for small volcanic aerosols.

Acknowledgments. Libraries supplying most of the older materials were the Columbia University Libraries, New York Public Library, American Museum of Natural History Library, Biblioteka Narodowa (Warsaw), Bibliothèque Interuniversitaire (Montpellier), Bibliothèque Nationale (Paris), National Meteorological Library (Bracknell, England), and Swedish Meteorological and Hydrological Institute Library (Norrköping). This paper also owes much to the methodologies developed by C. G. Abbot, H. H. Kimball, F. E. Volz, and D. V. Hoyt. A detailed record of the Crête ice core acidities was kindly supplied by C. U. Hammer. Thanks finally go to J. E. Hansen, A. A. Lacis, M. I. Mishchenko, M. Sato, and two anonymous reviewers for their suggestions and to the NASA Climate Research Program for overall support.

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R. B. Stothers, Goddard Institute for Space Studies, NASA, 2880 Broadway, New York, NY 10025.

(Received June 16, 1995; revised September 18, 1995; accepted September 18, 1995.)